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ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND ABERD--ETC F/G 20/3
SYMMETRY IN ELECTROMAGNETIC SCATTERING FROM A CLUSTER OF SPHERE--ETC(U)
AUG 80 O I SINDONI, F BORGHESE, P DENTI

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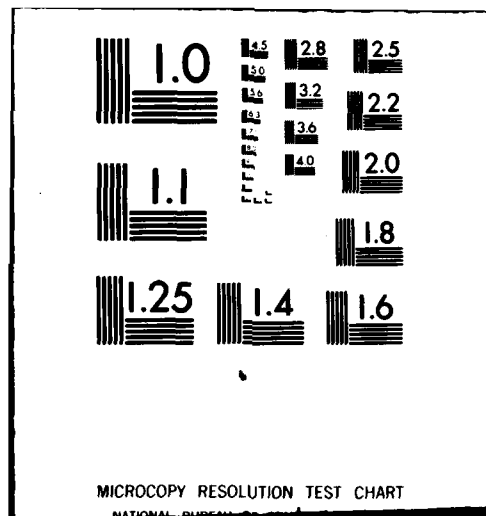
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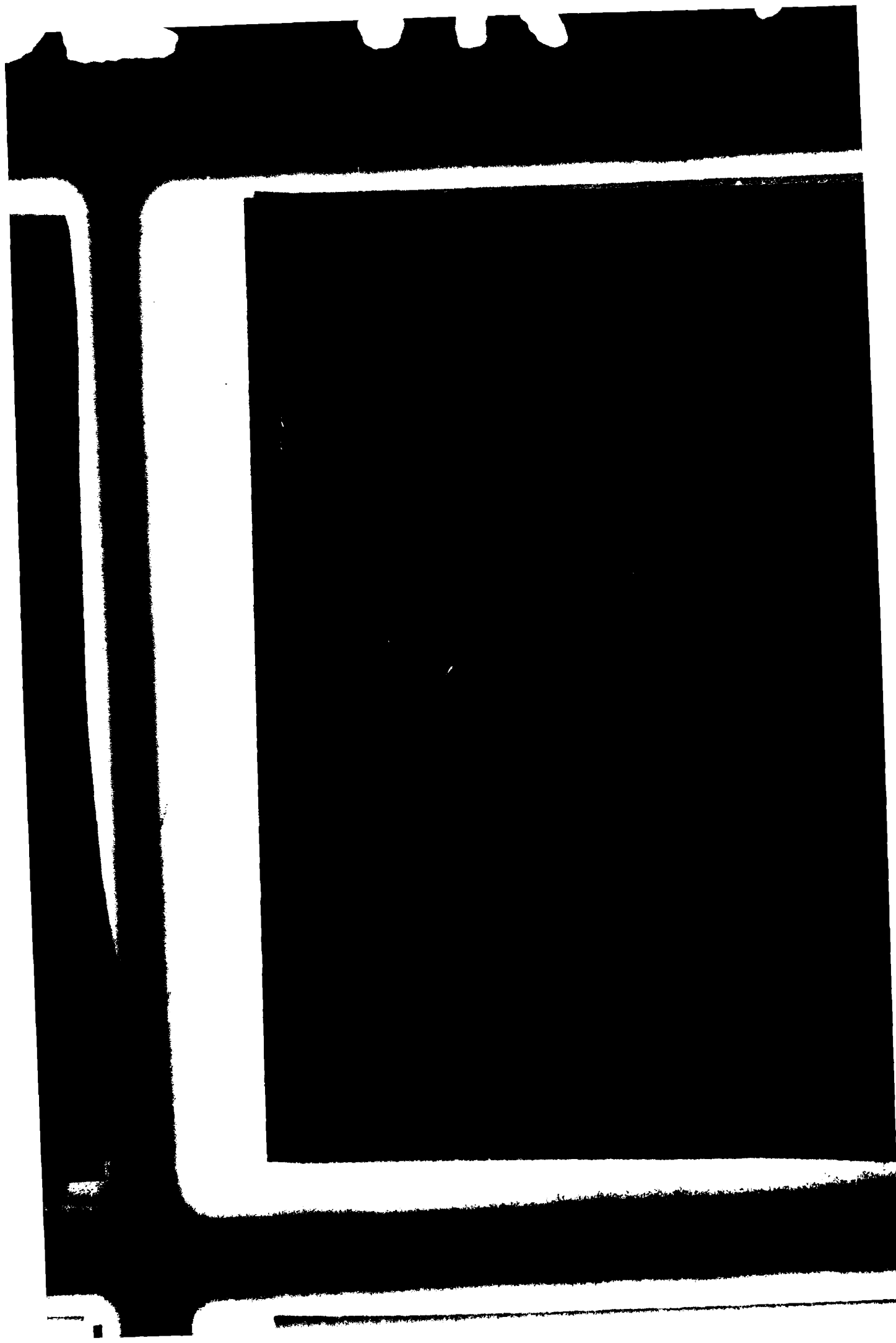
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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER ARCSL-TR-79066	2. GOVT ACCESSION NO. ✓ AD-A089648	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) SYMMETRY IN ELECTROMAGNETIC SCATTERING FROM A CLUSTER OF SPHERES		5. TYPE OF REPORT & PERIOD COVERED Technical Report April 1978-June 1979
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) O. I. Sindoni F. Borghese P. Denti G. Toscano		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS Commander/Director, Chemical Systems Laboratory DRDAR-CLB-PS Aberdeen Proving Ground, Maryland 21010		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS Project 1T161102A71A, Task 5
11. CONTROLLING OFFICE NAME AND ADDRESS Commander/Director, Chemical Systems Laboratory DRDAR-CLJ-R Aberdeen Proving Ground, Maryland 21010		12. REPORT DATE August 1980
		13. NUMBER OF PAGES 19
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report) UNCLASSIFIED
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE NA
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES Smoke/aerosol research		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Electromagnetic scattering Mie scattering Clusters Multiple scattering Debye potentials Cross section Refractive index Aerosols		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) In this study, a method for calculating the scattering properties of a cluster of dielectric spheres was developed and group theory was applied to simplify the calculations. The scattered field as well as the scattering and absorption cross sections can be computed without any restriction on the angle of incidence of the light or on the radii and refractive indices of the spheres in the cluster.		

PREFACE

The work described in this report was authorized under Project 1T161102A71A, Task 5, Aerosol/Obsecration Science. The work was started in April 1978 and completed in June 1979.

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Acknowledgment

This joint research between the Chemical Systems Laboratory and the University of Messina was made possible with support from the US Army European Research Office. Their cooperation is gratefully acknowledged.

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SYMMETRY IN ELECTROMAGNETIC SCATTERING FROM A CLUSTER OF SPHERES

I. INTRODUCTION.

This paper is a sequel to one we published in Applied Optics.¹ Henceforth, the previous article will be referred to as Part I. We shall briefly describe the subject as it was in Part I and then extend it here.

As stated in Part I, we are investigating the problem of electromagnetic scattering by particles of arbitrary shape. We have built a model scatterer, assuming for computational simplicity, to know the complex refractive indices and radii of the cluster of homogeneous nonmagnetic spheres of which it consists. The geometry of the cluster is rigid and we assume that the incident wave is a circularly polarized plane wave of wave-vector \mathbf{k} and frequency ω . Such an arrangement, where in the cluster the spheres are homogeneous and nonmagnetic, allowed us to transform the vector problem (the scattering problem has an intrinsic vector nature) into a pair of scalar ones using the Debye potentials to describe the electromagnetic field. In this way, we have simplified the problem and have been able to compute the absorption, scattering, and total cross sections.

The computations of the cross sections of the cluster and, therefore, the convergence of the scattered fields require solution of rather big systems whose order is given by $N(\ell_{\max} + 1)^2$, N being the number of the spheres in the cluster, and ℓ_{\max} the highest value of ℓ that is included in the expansions of Debye potentials. If, however, the cluster has a symmetry group, the above systems can be factorized through group-theory techniques similar to those used in quantum-mechanical problems. It is shown that the application of group theory to scattering produces a nonhomogeneous system of equations rather than secular determinants. These nonhomogeneous equations more closely represent real-world scattering by particles than is generally obtained.

II. THE SCATTERING EQUATIONS FOR GENERAL DIRECTION OF INCIDENCE.

In Part I, it was assumed that the incident wave propagated along the positive z -axis, the direction of incidence being then changed by rotating the cluster as a whole. For our present purposes, we shall refer the cluster to a fixed system of axes and choose the direction of incidence through the direction cosines of \mathbf{k} , the propagation vector of the wave. A straightforward calculation along the lines sketched by Jackson² shows that for a circularly polarized plane wave of wave-vector \mathbf{k} :

$$\begin{aligned} \mathbf{E}^{\pm}(\mathbf{r}) &= (\mathbf{e}_1 \pm i\mathbf{e}_2) \exp(i\mathbf{k} \cdot \mathbf{r}) = \\ 4\pi \sum_{\ell m} i^{\ell} (\mathbf{e}_1 \pm i\mathbf{e}_2) \cdot \mathbf{X}_{\ell m}^{\circ}(\hat{\mathbf{r}}) &\left[\mathbf{X}_{\ell m}(\mathbf{r}) j_{\ell}(kr) \pm \frac{1}{k} \Delta \mathbf{x} \cdot \mathbf{X}_{\ell m}(\mathbf{r}) j_{\ell}(kr) \right] \end{aligned} \quad (1a)$$

$$\mathbf{B}^{\pm}(\mathbf{r}) = \mp i \mathbf{E}^{\pm}(\mathbf{r}) \quad (1b)$$

where \underline{e}_1 and \underline{e}_2 are unit vectors orthogonal to \underline{k} and to each other and $\chi_{\ell m}(\hat{\underline{r}})$ and $\chi_{\ell m}(\hat{\underline{k}})$ are vector spherical harmonics defined as:

$$\chi_{\ell m}(\hat{\underline{r}}) = \frac{-i \underline{k} \times \nabla}{\sqrt{\ell(\ell+1)}} Y_{\ell m}(\hat{\underline{r}}), \quad \chi_{\ell m}(\hat{\underline{k}}) = \frac{-i \underline{k} \times \nabla}{\sqrt{\ell(\ell+1)}} Y_{\ell m}(\hat{\underline{k}}) \quad (2)$$

The Debye potentials for the fields (1) are then (Newton³)

$$\psi_0^\pm(\underline{r}) = \frac{1}{k} \sum_{\ell m} i^2 d_\ell Q_{\ell m}^*(\hat{\underline{k}}) j_\ell(kr) Y_{\ell m}(\hat{\underline{r}}) \quad (3a)$$

$$\phi_0^\pm(\underline{r}) = \pm \psi_0^\pm(\underline{r}) \quad (3b)$$

with

$$d_\ell = 4\pi [\ell(\ell+1)]^{-1/2}, \quad Q_{\ell m}^*(\hat{\underline{k}}) = (\underline{e}_1 \pm i \underline{e}_2) \cdot \chi_{\ell m}^*(\hat{\underline{k}})$$

When the above expressions for ϕ_0^\pm and ψ_0^\pm are introduced into the formalism developed in Part I, the final systems of equations which determine the coefficients $A_{\ell m}^\alpha$, $B_{\ell m}^\alpha$ become

$$\sum_{\beta} \sum_{\ell' m'} \left\{ \delta_{\alpha\beta} \delta_{\ell\ell'} \delta_{mm'} \left[S_{\ell'}^\beta \right]^{-1} + G_{\ell m; \ell' m'}^{\alpha\beta} \right\} A_{\ell' m'}^\beta = -2e^{\underline{k} \cdot \underline{R}_\alpha} Q_{\ell m}^*(\hat{\underline{k}}) \quad (4a)$$

$$\sum_{\beta} \sum_{\ell' m'} \left\{ \delta_{\alpha\beta} \delta_{\ell\ell'} \delta_{mm'} \left[T_{\ell'}^\beta \right]^{-1} + G_{\ell m; \ell' m'}^{\alpha\beta} \right\} B_{\ell' m'}^\beta = -2e^{\underline{k} \cdot \underline{R}_\alpha} Q_{\ell m}^*(\hat{\underline{k}}) \quad (4b)$$

where S_ℓ^α , T_ℓ^α and $G_{\ell m; \ell' m'}^{\alpha\beta}$ are still defined as before.

III. SYMMETRIZATION OF DEBYE POTENTIALS.

The symmetrization of the systems (4) requires us to symmetrize only ϕ_Π and ψ_Π , the Debye potentials in the intersphere region. The expansion coefficients of ϕ_I and ψ_I , the potentials within the spheres, cancel out of both sides of the resulting equations. It is convenient to partition the cluster in subsets of spheres which are related to each other by the symmetry operations and to indicate the vector position of the α -th sphere in the σ -th subset by $\underline{R}_\alpha^\sigma$, while $\underline{r}_\alpha^\sigma = \underline{r} - \underline{R}_\alpha^\sigma$. The expansion of ϕ_Π in terms of symmetry-adapted combinations of Helmholtz solid harmonics is:

$$\phi_\Pi(\underline{r}) = \pm \frac{1}{2k} \sum_{\mu p} \sum_{\ell} i^2 d_\ell \left[\sum_{\sigma n} A_{n\ell}^{\mu p \sigma} H_{n\ell}^{\mu p \sigma}(\underline{r}) + 2 \sum_{\ell m} Q_{\ell m}^*(\hat{\underline{k}}) F_{m, \ell\ell}^{\mu p} J_{\ell\ell}^{\mu p}(\underline{r}) \right] \quad (6)$$

where we use the definitions

$$H_{nl}^{\mu p \sigma}(\mathbf{r}) = \sum_{\alpha m} a_{nlm}^{\mu p \sigma \alpha} h_l^{(1)}(\mathbf{kr}_\alpha^\sigma) Y_{lm}(\hat{\mathbf{r}}_\alpha^\sigma) \quad (7)$$

for the combinations of solid harmonics centered at the various sites of the σ -th subset and

$$J_{nl}^{\mu p}(\mathbf{r}) = \sum_m b_{nlm}^{\mu p} j_l(kr) Y_{lm}(\hat{\mathbf{r}}) \quad (8)$$

for the combinations of functions centered at the origin.

In the preceding equations, the superscripts μ, p refer to the p -th row of the μ -th irreducible representation. Note that $H_{nl}^{\mu_1 p} = H_{nl}^{\mu_2 p}$, $J_{nl}^{\mu_1 p} = J_{nl}^{\mu_2 p}$ so that a given irreducible representation may appear more than once for a given l . The symmetrization coefficients, $a_{nlm}^{\mu p \sigma \alpha}$ and $b_{nlm}^{\mu p}$, are easily obtained by standard techniques^{4, 5} through the use of the well-known transformation formulas for spherical harmonics.⁶ A quite analogous expansion can be written for ψ_Π , the coefficients being indicated by $B_{nl}^{\mu p \sigma}$. A comparison of the symmetrized expansions with their unsymmetrized counterparts yields the equations

$$A_{lm}^{\alpha \sigma} = \sum_{\mu p m} A_{nl}^{\mu p \sigma} a_{nlm}^{\mu p \sigma \alpha}; B_{lm}^{\alpha \sigma} = \sum_{\mu p m} B_{nl}^{\mu p \sigma} a_{nlm}^{\mu p \sigma \alpha} \quad (9)$$

which allow us to calculate the cross sections whose expressions involve the $A_{lm}^{\alpha \sigma}$'s and the $B_{lm}^{\alpha \sigma}$'s.

IV. FACTORIZATION OF THE SYSTEMS FOR THE COEFFICIENTS.

The equation for the coefficients $A_{nl}^{\mu p \sigma}$ and $B_{nl}^{\mu p \sigma}$ are obtained by imposing on ϕ and ψ the well-known boundary conditions (Newton³) on the surface of each sphere. To do this, we first put ϕ_Π and ψ_Π in a form involving only Helmholtz solid harmonics centered at a single site, say \mathbf{R}_α^σ , through the same addition formulas^{7, 8} we have already used for the unsymmetrized case. The result for ϕ_Π is:

$$\begin{aligned} \phi_\Pi(\mathbf{r}_\alpha^\sigma) = & \pm \frac{1}{2k} \sum_{\mu p} \sum_{lm} i^l d_l Y_{lm}(\hat{\mathbf{r}}_\alpha^\sigma) \left\{ \sum_n \left[A_{n'l}^{\mu p \sigma} a_{n'l m}^{\mu p \sigma \alpha} h_l^{(1)}(\mathbf{kr}_\alpha^\sigma) + \right. \right. \\ & \left. \sum_{l'm', \beta \tau} G_{lm; l'm'}^{\alpha \sigma, \beta \tau}(\mathbf{R}_{\alpha \beta}^{\sigma \tau}) a_{n'l'm'}^{\mu p \tau \beta} A_{n'l}^{\mu p \tau} j_l(kr_\alpha^\sigma) \right] + \\ & \left. 2 \sum_{\bar{n}} \sum_{\bar{l} \bar{m}'} P_{\bar{n} \bar{l}}^{\mu p} \bar{G}_{\bar{l} m; l'm'}^{\alpha \sigma}(-\mathbf{R}_\alpha^\sigma) b_{\bar{n} \bar{l} m'}^{\mu p} j_{\bar{l}}(kr_\alpha^\sigma) \right\} \quad (10) \end{aligned}$$

where we put

$$P_{\bar{n} \bar{l}}^{\mu p} = \sum_{m''} Q_{\bar{l}, m''}^\sigma \otimes F_{m'', \bar{n} \bar{l}}^{\mu p}$$

and:

$$\tilde{G}_{\ell m; \ell' m'}^{\alpha \sigma}(-\mathbf{R}_\alpha^\sigma) = 4\pi \frac{d_{\ell'}}{d_\ell} \sum_L I_L(\ell m; \ell' m') i^{-L} Y_{L, m-m'}^*(\hat{\mathbf{R}}_\alpha^\sigma) j_L(k R_\alpha^\sigma) \quad (11)$$

When we impose the required boundary conditions on ϕ at the surface of the α -th sphere in the σ -th subset we get for each μ, p an equation of the form:

$$\sum_{\tau \beta} \sum_{n'} \sum_{\ell' m'} \left\{ \delta_{\sigma \tau} \delta_{\alpha \beta} \delta_{\ell \ell'} \delta_{m m'} [S_{\ell'}^\tau]^{-1} + G_{\ell m; \ell' m'}^{\alpha \sigma, \beta \tau} \left\{ a_{n' \ell' m'}^{\mu \tau \beta} A_{n' \ell'}^{\mu \tau} \right\} \right. \\ \left. - 2 \sum_{\bar{n}} \sum_{\alpha \ell' m'} P_{\bar{n} \ell'}^{\mu p} \tilde{G}_{\ell m; \ell' m'}^{\alpha \sigma} b_{\bar{n} \ell' m'}^{\mu p} \right\} = \quad (12)$$

We notice that $S_{\ell'}^\tau$ should be written as $S_{\ell'}^{\beta \tau}$ but, since it is actually independent of the site within each subset, the superscript β has accordingly been dropped. Now, if equation 12 is multiplied by $(a_{n \ell m}^{\mu p \sigma \alpha})^*$ and summed over α and m and fixing the superscripts μ, p , and dropping them from the equation, we get

$$\sum_{n' \ell'} \sum_{\tau} \left\{ \delta_{\sigma \tau} \delta_{\ell \ell'} \delta_{m m'} [S_{\ell'}^\tau]^{-1} + G_{\ell n; \ell' n'}^{\sigma \tau} \right\} A_{n' \ell'}^\tau = -2 \sum_{\bar{n} \ell'} P_{\bar{n} \ell'} \tilde{G}_{\ell n; \ell' \bar{n}}^\sigma \quad (13)$$

Equation 13 is obtained by applying the following techniques:

$$G_{\ell n; \ell' n'}^{\sigma \tau} = \sum_{\alpha \beta} \sum_{m m'} \left(a_{n \ell m}^{\sigma \alpha} \right)^* G_{\ell m; \ell' m'}^{\alpha \sigma, \beta \tau} a_{n' \ell' m'}^{\tau \beta}$$

and

$$\tilde{G}_{\ell n; \ell' \bar{n}}^\sigma = \sum_{\alpha} \sum_{m m'} \left(a_{n \ell m}^{\sigma \alpha} \right)^* \tilde{G}_{\ell m; \ell' m'}^{\alpha \sigma} b_{\bar{n} \ell' m'}$$

and where we have taken into account that, if the irreducible representations are chosen properly (Altmann and Bradley⁹ and Diamond⁶), the symmetrization coefficients satisfy

$$\sum_{\alpha m} \left(a_{n \ell m}^{\mu p \sigma \alpha} \right)^* a_{n' \ell' m}^{\mu p \sigma \alpha} = \delta_{n n'} \quad (14)$$

so that

$$\delta_{\sigma \tau} \delta_{\ell \ell'} \sum_{\alpha m} \delta_{\alpha \beta} \delta_{m m'} \left(a_{n \ell m}^{\sigma \alpha} \right)^* [S_{\ell'}^\tau]^{-1} a_{n' \ell' m'}^{\tau \beta} = \delta_{\sigma \tau} \delta_{\ell \ell'} [S_{\ell'}^\tau]^{-1}$$

Equations quite analogous to equation 13 hold true for the B_{nl}^{τ} 's.

We recall that (although never explicitly indicated) the coefficients $A_{nl}^{\mu\nu\sigma}$ and $B_{nl}^{\mu\nu\sigma}$ depend on both the polarization and on the direction cosines of \mathbf{k} . Furthermore, once the A 's and B 's have been computed, the C 's and D 's, the expansion coefficients of the symmetrized Debye potentials within the spheres, are calculated through equation 16 of Part I which are still valid on account of equations 14 and 9.

V. THE CROSS SECTIONS.

The right-hand side of equation 13 involves the quantities $\tilde{G}_{lm;l'm'}^{\sigma}$ which are easily identified as the symmetrized matrix elements, in the site and angular momentum representation, of the free-space propagator for plane waves.¹⁰ Doing this not only simplifies the expression of the incident wave in symmetrized form but also allows the calculation of the cross section without any approximation other than the truncation of the l -expansion of Debye potentials. Indeed the scattered potentials are given by:

$$\psi_s(\mathbf{r}) = \frac{1}{2k} \sum_{\alpha\sigma} \sum_{lm} i^l d_l B_{lm}^{\alpha\sigma} Y_{lm} \left(\hat{\mathbf{r}}_a \right) h_l^{(1)} \left(kr_a^\sigma \right) \quad (15a)$$

$$\phi_s(\mathbf{r}) = \pm \frac{1}{2k} \sum_{\alpha\sigma} \sum_{lm} i^l d_l A_{lm}^{\alpha\sigma} Y_{lm} \left(\hat{\mathbf{r}}_a \right) h_l^{(1)} \left(kr_a^\sigma \right) \quad (15b)$$

and, as the potentials of the incident wave involve only Helmholtz harmonics centered at the origin, it is convenient to use again the addition formulas of Nozawa⁷ to transform equation 15 into the form:

$$\psi_s(\mathbf{r}) = - \frac{1}{2k} \sum_{\alpha\sigma} \sum_{lm} \sum_{l'm'} i^l d_l B_{l'm'}^{\alpha\sigma} \tilde{G}_{lm;l'm'}^{\alpha\sigma} \left(\frac{\mathbf{R}^\sigma}{\alpha} \right) Y_{lm}(\hat{\mathbf{r}}) h_l^{(1)}(kr) \quad (16a)$$

$$\phi_s(\mathbf{r}) = \pm \frac{1}{2k} \sum_{\alpha\sigma} \sum_{lm} \sum_{l'm'} i^l d_l A_{l'm'}^{\alpha\sigma} \tilde{G}_{lm;l'm'}^{\alpha\sigma} \left(\frac{\mathbf{R}^\sigma}{\alpha} \right) Y_{lm}(\hat{\mathbf{r}}) h_l^{(1)}(kr) \quad (16b)$$

We notice, incidentally, that when $kR_a^\sigma \ll 1$, equation 16 reduces to equation 17 of Part I for on account of the small-argument behavior of the spherical Bessel functions

$$\lim_{kR_a^\sigma \rightarrow 0} \tilde{G}_{lm;l'm'}^{\alpha\sigma} = \delta_{ll'} \delta_{mm'}$$

By defining

$$\bar{A}_{lm}^{\alpha\sigma} = \sum_{l'm'} \tilde{G}_{lm;l'm'}^{\alpha\sigma} A_{l'm'}^{\alpha\sigma}, \quad \bar{B}_{lm}^{\alpha\sigma} = \sum_{l'm'} \tilde{G}_{lm;l'm'}^{\alpha\sigma} B_{l'm'}^{\alpha\sigma} \quad (17)$$

we get for the cross sections:

$$\sigma_x = \frac{2\pi^2}{k^2} \sum_{\ell m} \sum_{\alpha\sigma} \sum_{\beta\tau} \left[\bar{A}_{\ell m}^{\alpha\sigma*} \bar{A}_{\ell m}^{\beta\tau} + \bar{B}_{\ell m}^{\alpha\sigma*} \bar{B}_{\ell m}^{\beta\tau} \right] \quad (18)$$

$$\sigma_{\text{abs}} = \frac{2\pi^2}{k^2} \sum_{\ell m} \sum_{\alpha\sigma} \sum_{\beta\tau} \left[2Q_{\ell m}^* Q_{\ell m} - \left(\bar{A}_{\ell m}^{\alpha\sigma*} + Q_{\ell m} \right) \left(\bar{A}_{\ell m}^{\beta\tau} + Q_{\ell m}^* \right) - \left(\bar{B}_{\ell m}^{\alpha\sigma*} + Q_{\ell m} \right) \left(\bar{B}_{\ell m}^{\beta\tau} + Q_{\ell m}^* \right) \right] \quad (19)$$

and:

$$\sigma_{\text{tot}} = \frac{4\pi^2}{k^2} \sum_{\ell m} \sum_{\alpha\sigma} \text{Re} \left[Q_{\ell m} \left(\bar{A}_{\ell m}^{\alpha\sigma} + \bar{B}_{\ell m}^{\alpha\sigma} \right) \right] \quad (20)$$

VI. DISCUSSION.

The procedure described in the preceding sections does not differ from that customarily used to factorize secular determinants, except that the inhomogeneity of our equations forces us to solve *all* the systems arising from equation 4. Indeed, the quantities S_p^β , T_p^β , on the left-hand side of equation 4 are the matrix elements, in the site and angular momentum representation, of the $\hat{\tau} + \hat{G}$, the $\hat{\tau}$ -operator accounting for the scattering power of a single sphere¹¹ and \hat{G} being the free space propagator for outgoing spherical waves.¹² As the operator $\hat{\tau} + \hat{G}$ is invariant under the symmetry group of the cluster, its symmetrized matrix elements, i.e., the quantities within braces in equation 13, are actually independent of the row index, p . On the other hand, the potentials of the incident wave on the right-hand side of equation 4 are not invariant under the symmetry group. According to general theorems, they have been decomposed into parts belonging to the rows of the irreducible representations, but these symmetrized parts do actually depend on the row index. However, because of the above-discussed independence of p on the left-hand side of equation 13, very little extra computational work is required to solve all systems belonging to a multidimensional representation. From a computational point of view, most of the computing time is, in fact, spent to invert the matrix of $\hat{\tau} + \hat{G}$ and, as the required time grows more quickly than the order of the matrix, the factorization procedure is always very useful.

As an example, let us consider a tetrahedral cluster with CH_4 structure (symmetry group T_d). By including terms up to $\ell_{\text{max}} = 2$, we get two unsymmetrized systems of order 45. However, by using symmetrized expansions, each one of the above systems splits into one A_2 system of order 3, one A_1 of order 1, two E of order 4, three F_1 of order 6, and three F_2 of order 5. The usefulness of the factorization through group theory needs no further comment.

VII. SUMMARY.

The work reported here and in our previous papers has led to a unique approach to calculating the optical properties of clusters of molecules. It now appears possible to compute

extinction of a particle knowing only its chemical composition. This is a radical departure from the Mie theory, yet the solution converges to Mie results for a single sphere. The theory developed here will now be tested using the $X \alpha - \omega$, scattered wave computer program developed by Johnson, *et al.* at IBM and modified by the authors for this study. The initial material to be investigated will be sulfuric acid, a material which is optically well characterized. This validation effort will be the subject of a subsequent technical report.

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